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ISOLDE and Neutron Time-of-Flight Experiments Committee

Semiconductor Spectroscopy with Short Lived Isotopes

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Summary

Electronic properties of semiconductors are extremely sensitive to defects and impurities that have localized electronic states with energy levels in the band gap of the semiconductor. Spectroscopic techniques used in semiconductor physics like photoluminescence (PL), deep level transient spectroscopy (DLTS), or Hall effect, that are able to detect and characterize band gap states do not reveal direct information about their microscopic origin. To overcome this chemical “blindness” of the electrical and optical methods the present approach is to use radioactive isotopes as a tracer. To enable the use of isotopes with half-lives down to about 1 h, we propose to perform PL, DLTS, and Hall effect measurement “on site” in a laboratory near the ISOLDE hall. We will shortly review the present results using long-lived isotopes to identify defect states in Si, III-V semiconductors, and SiC and propose new experiments using short-lived isotopes which require that the measurements must be performed “on-site”:

- Hydrogen and lithium interaction with gold and platinum in silicon
- Identification of transition metal defects in silicon
- Identification of Acceptor and donor states in GaN
- Creation and identification of anti-site defects in GaAs and GaN
- Alkali and alkaline earth defects in Si and Ge
- Identification of acceptor states in II-VI semiconductors
- Identification of donor and acceptor centers in ZnO
- Deep states of group III acceptors in SiC

1. Introduction

Electronic properties of semiconductors are extremely sensitive to defects and impurities. Small concentrations of 10^{-12} cm^{-3} or less can drastically influence the type and magnitude of electronic conductivity or the lifetime of charge carriers. This is the case, if the impurity or defect in question gives rise to a localized electronic state having an energy level within the band gap of the semiconductor. Because of the semiconductor's high sensitivity to low defect concentrations, a precise knowledge of impurity related band gap states is highly desirable, either to understand a given crystal and its defects or to tailor its properties by intentional doping. The intentional, spatially structured modification of the material's properties by incorporation of suitable impurities (‘doping’) and the prevention of the unintentional incorporation of defects during the many processing steps necessary to build a device is the very basis of modern semiconductor electronics.

Progress in semiconductor technology is driven by two requirements: Developing new materials with unique optical or electrical features and reducing the size of the individual constituents of an integrated device. These requirements demand a thorough understanding and control of defects in these materials; both of intrinsic defects, such as vacancies, self-interstitials, or anti-sites, and of extrinsic defects, such as dopants and impurity atoms. As a consequence, a strong effort is currently devoted to the investigation of defects and the electrical activation of dopant atoms.

Generally, spectroscopic techniques used in semiconductor physics like photoluminescence (PL) or deep level transient spectroscopy (DLTS) which are able to detect and characterize band gap states do not reveal direct information about their microscopic origin. On the other hand, structurally or chemically sensitive methods, like magnetic resonance techniques, Mößbauer spectroscopy, perturbed angular correlation spectroscopy, or emission channeling, normally do not disclose the energetic position of a state within the band gap. To overcome this chemical “blindness” of the electrical and optical methods the present approach is to use radioactive isotopes as a tracer. Because of the characteristic concentration change according to the nuclear decay law, their involvement in the formation of electronic band gap states can be confirmed or denied definitely by several subsequent spectroscopic measurements during the elemental transmutation. Band gap states related to either the parent or the daughter isotope are uniquely identified by a decreasing or increasing concentration, respectively.

In principle all radioactive isotopes can be used for this type of experiments. However, if the measurements take place at the laboratories outside of CERN the shortest feasible half-lives are at least one day. This is due to the fact that for the evaluation of the experiment it is crucial to observe the time evolution of the spectra from the starting point, the implantation. If one wants to observe the increase of the signal of the daughter isotope the fastest change occurs in the first half-life after the implantation of the parent isotope.

To overcome these limitations we propose to perform photoluminescence (PL), deep level transient spectroscopy (DLTS) and Hall effect measurement on site in a laboratory near the ISOLDE hall. If only the time for sample preparation such as post implantation annealing, preparation of electrical contacts, and for mounting the sample into the experimental setup is needed, the use of isotopes with half-lives as short as hours is possible.

This opens up a number of experiments with elements where no long-lived isotopes exist, especially in the region of lighter elements. The use of short-lived isotopes also makes sense if there exist long-lived isotopes of the same element. The short half-life has the advantage that several experiments can be done during one beam time increasing the efficiency of the experiment, and that it is easier to keep all measurement conditions stable during a series of measurements. When using long lived isotopes with measurement periods up to months, keeping boundary conditions stable and reproducible can cause serious problems. This includes storing of samples between the measurements, reproducible illumination of the same part of the sample by laser light, and last but not least the possible degradation of the sample itself by diffusion or defect reactions etc.

Since the early 90s, a series of successful experiments using radioactive DLTS (IS 321, IS 325, IS 345) and PL (IS 357, IS 369) has been started using the radioactive ion beams produced by ISOLDE. These experiments, up to now, only used radioactive isotopes with a half-life of at least one day, so that the actual measurements could be performed at the respective home institutes. It is the aim of this proposal to open the possibility to execute DLTS, PL and Hall effect measurements on-site at CERN and therefore extend the possibilities for these techniques significantly by closing the gap for condensed matter spectroscopy experiments at ISOLDE between on-line experiments and just collecting long-lived isotopes to perform various experiments at the respective home institutes. In the following we will illustrate a few such “on-site” experiments using DLTS, PL spectroscopy and Hall effect measurements. Both the proposed experiments in semiconductor physics and the used experimental techniques are only a small selection of the possible work, which could be done in condensed matter studies if the prerequisites for this type of experiments exist. During the preparation of this proposal almost all groups from solid state physics and life sciences working at ISOLDE have expressed their interest in such type of experiments and additional experiments may be presented in future proposals.

2. Experimental methods

Deep Level Transient Spectroscopy (DLTS):

Among the techniques available to characterize band gap states, the deep level transient spectroscopy (DLTS) is one of the most powerful and well-established standard techniques. In its standard version, it is a purely electrical measuring technique using space charge junctions like Schottky or pn-diodes on the material to be investigated. DLTS is very sensitive (detecting a few as 1000 defects in a junction) and provides quantitative information about the energetic level position, capture cross section and the concentration of band gap states but not on the chemical identity of a defect. To overcome this “chemical blindness”, DLTS has been used on samples doped with radioactive tracers. Starting with the identification of Au related deep levels in Si by implantation of ^{195}Au→Pt at ISOLDE [1], various experiments have been performed with radioactive isotopes implanted off-line or by recoil implantation [2].

Some groups (Jena and Berlin/Dresden) have used radiotracer-DLTS for several years in silicon [3,4,5,6] and in the “high-temperature semiconductor” silicon carbide with radioactive isotopes of the elements Ti, V, Cr, Co, Ga, As, In, and Ta [7].

The number of suitable isotopes is large, since there is one single requirement only: a reasonable half-life. The half-lives used up to now are between 3.08 h [8] and 183 d [1]. The

lower limit is set by the time needed for implantation, annealing and contact preparation and is close to the 3 h already used. Half-lives in the order of a few days or less generally require the sample preparation and measurement to be performed on-site at the implantation facility.

Photoluminescence Spectroscopy (PL):

The most common means employed for generating luminescence in semiconductors is to use an optical pump (usually a laser) to produce electrons and holes in the semiconductor. When these carriers become bound at defects or impurities and recombine with holes or electrons respectively, photons are emitted of energies correlated to the energy level of the defect in the band gap. The spectrum of the recombination energies, the vibrational side bands, and temperature dependence of the intensities, etc. - contains a wealth information on the defect or impurity. The term photoluminescence (PL) is used to denote such optically generated luminescence. PL is employed widely in all classes of semiconductor research and is recognized as one of the most important and powerful techniques available [9]. The principal advantage is, in addition to the usual advantages pertaining to spectroscopy, the potential for very high sensitivity. For semiconductors, the penetration depth of the laser radiation and the diffusion length of electrons and holes are generally of the order of 100 nm, often less. A typical laser beam has a cross-sectional area of ~1 mm^2. Accordingly, the effective sample volume for PL is of the order of 10^{-4} cm^3. Depending on the recombination process, the detection limit can be as low as 10^{13} cm^{-3} in favorable cases, so that a total of 10^9 atoms can be sufficient to produce detectable PL signals. This can be lowered further by focusing the laser to a spot, for example. To get the full benefits from PL it is usually necessary to operate at liquid helium temperatures to freeze out all thermally excited charge carriers. At these temperatures even weakly bound electrons and holes remain bound at defects/impurities so that the optical transitions occur and vibrational (phononic) effects are minimized. One consequence of this high sensitivity is the capability of detecting trace contaminants in nominally intrinsic semiconductors.

However, the assignment to a particular defect is often a puzzle. In semiconductors with residual impurities below the threshold concentration for PL detection intentional doping and its correlation to the intensity of PL transitions is used to identify the chemical nature of defect levels. In this way many defect levels in Si, Ge, and GaAs have been identified during the last decades. However, in other semiconductors, like InP, GaN or the II-VI compounds, which are of growing interest for opto-electronic applications, many defect levels are still not identified, due to the difficulty to grow high purity crystals. In general, an unambiguous chemical identification of a defect level is provided only by the observation of an element specific property, like the isotope mass, the nuclear spin, or the isotope abundance. If light impurities are involved in the defect one can observe the isotope shift of no-phonon lines or local mode phonon replicas [10]. Another possibility exists in the determination of the hyperfine interaction by electron paramagnetic resonance (EPR) or electron-nuclear double resonance (ENDOR) experiments where selective photo-excitation or photo-ionization of a paramagnetic level helps to determine the defect level [11]. These two methods, however, require a paramagnetic level, suitable nuclear properties of the involved elements, and a control over the Fermi-level for selective photo-excitation. Due to these limitations any additional method to identify defect levels is highly welcome.

Therefore, the use of the nuclear lifetime of a radioactive isotope undergoing a chemical transmutation has become a most versatile tool to identify defect levels. If the level is caused

by a defect in which the parent or daughter isotope is involved the concentration of that defect will change with the characteristic time constant of the radioactive decay. The first successful application of this technique was demonstrated in 1965 by I. Broser and K.-H. Franke [12] who doped ZnS homogeneously with radioactive ^{65}Zn by neutron irradiation. Due to the decay of ^{65}Zn to ^{65}Cu they have been able to determine the defect level of Cu in ZnS. It took about 25 years until this type of PL experiment was reinvented by the Konstanz and Dublin groups to identify defect states in Si [13,14] and GaAs [15]. In all these experiments ion implantation was used for doping with radioactive isotopes. This offers a lot more possibilities for doping with radioactive dopants than neutron irradiation does. Furthermore, the impurities in the neutron-irradiated samples can give rise to unwanted production of radioactive isotopes. By ion implantation only the wanted dopant is introduced due to its mass selectivity.

Hall effect:

The most obvious quantity characterizing a semiconductor is its electrical conductivity which is determined by the type of majority charge carriers present, their concentration, and their mobility. These quantities can be determined by measuring the specific conductivity and the Hall effect as function of temperature. The theoretical basis and the experimental realization is described in all introductory books on solid state physics; a more thorough discussion can be found in [16]. If the observed type and concentration of carriers is caused by a defect in which the parent or daughter isotope is involved the concentration of that defect will change with the characteristic time constant of the radioactive decay and therefore changes in conductivity and carrier concentration can be associated with a specific defect.

3. Physics Motivation and Proposed Experiments

3.1 Silicon and Germanium

All silicon crystals contain traces of impurity atoms, notably hydrogen and some metals of the first transition series (in addition to carbon and oxygen). These impurities produce a variety of single- and multi-atom defects in the crystals, and it is our aim to establish the experimental facts regarding these defects. The key facts to be established include: (i) the positions of the impurity levels in the semiconductor energy gap; (ii) the fine-structure in the ground state and excited state energy level manifolds; (iii) full chemical identification of the elements involved in each defect. Such fundamental information provides the foundation for theoretical models. Experience shows that, along with EPR, DLTS and PL are among the best tools for this purpose, and the results we have already achieved at ISOLDE confirm the value of these techniques when used in conjunction with radioactive isotopes for chemical identification [6,14,17].

H and Li interactions with Au and Pt:

The majority of our measurements to date have been focused on identifying the characteristic DLTS and PL spectra for isolated Au, isolated Pt and Au-Fe, and Pt-Fe centers. We have corrected several false assignments of PL spectra in the literature regarding Au and Pt-related defects, and we have re-opened the question of the occurrence of a midgap deep level due to Au in p-type silicon [14,17,18]. As an example, Fig. 1 shows the changes that occur in the PL spectrum of Si doped with ^{193}Au (17.7 h). Only the regions of interest are shown. The 735 meV and 1066 meV bands both decrease as the Au population decreases. The

777 meV band which grows in intensity is due to Pt. The line at 1066 meV has been earlier observed after diffusion of Fe and attributed to a FeB complex [19]. However, the corresponding PL intensity decays with the characteristic half-life $t_{1/2}$ = 17.7 h of ^{193}Au. This is a direct proof that Au has to be involved in this complex.

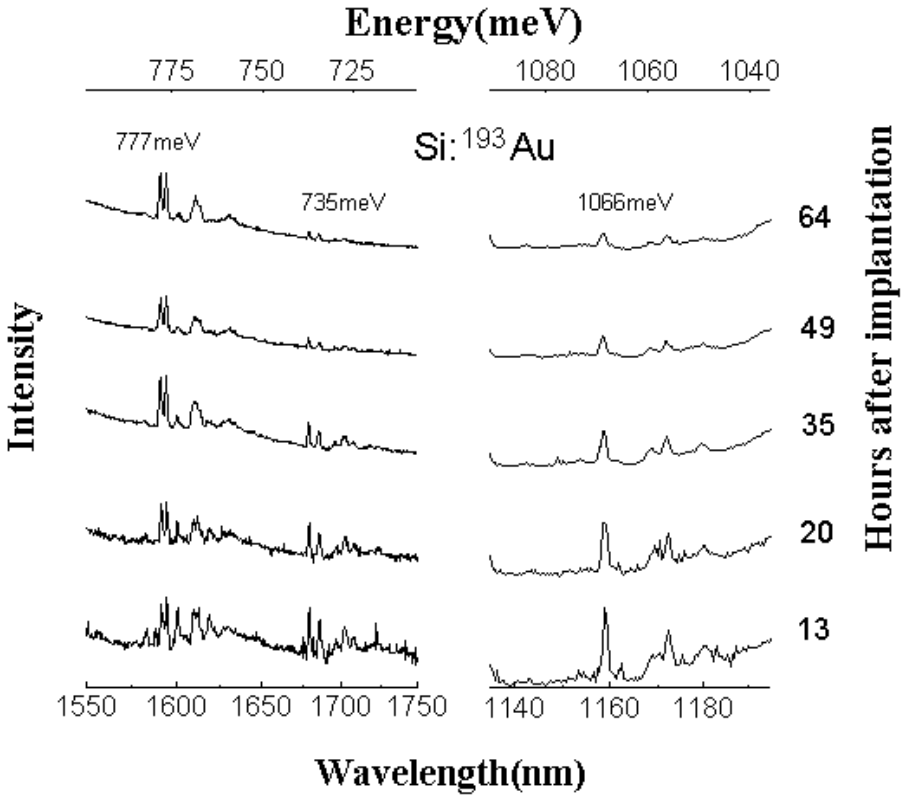

Fig. 1*: The changes that occur in the PL spectrum of Si:^{193}Au. Only the regions of interest are shown. The 735 meV and 1066 meV bands both decrease as the Au population decreases. The 777 meV band which grows in intensity is due to Pt.*

H and Li are described as the light impurities in silicon. These impurities are characterized by their tendency to interact with other impurities and to produce multi-atom H and Li clusters. IR absorption and EPR measurements provide some evidence for the occurrence of complex defects involving H and Li with Au and Pt [20,21]. However, some essential data, especially the correlation of the Au and Pt forms of some defects, have not been reported. Furthermore, a comprehensive model to account for the variety of defects produced has not been developed.

The experiments we propose involve the preparation of samples with radioactive Au which transforms to Pt. We have already produced samples (using stable isotopes) for which we observe the PL signatures believed to be due to Au-Li centers [22]. We have not been able to produce the Pt-Li counterpart using stable Pt isotopes for reasons that we do not know. We are very well placed to produce the Au-Li centers using radioactive ^{191}Au (3.2 h) and ^{193}Au (17.7 h) and to investigate the daughter Pt-Li centers, thereby obtaining direct comparisons of the Au and Pt forms of the defects. Such precise control cannot be achieved by preparing different samples with stable isotopes of the two elements. Both DLTS and PL, which have been successfully used in experiment IS 357, will be employed in the measurements. Preparatory experiments using stable isotopes are still required for the case of Au-H and Pt-H

studies. However, the sample preparation techniques are fully described in the literature [20] and we expect to be able to investigate this topic too in the course of the proposed work.

Identification of transition metal defect signatures for silicon:

Commercially available silicon crystals contain some metal impurities (such as Fe and Cu) at concentrations of typically 10^{14} cm^{-3}. The single biggest issue in all experimental investigations has been the chemical identity of the impurities present in the samples. The elements in the range Mn to Zn, especially Fe to Cu, have high diffusivities in silicon and a pronounced tendency to form precipitates or complexes and therefore are readily dispersed throughout silicon wafers during short exposures to high temperatures. Consequently, these are the impurities for which doubts about defect identifications are greatest. The development at ISOLDE of laser ionization techniques for the production of some metal isotopes provides an excellent opportunity to study the critical first transition series. Our priority experiments will be focused on Si:Cu since Cu, one of the ubiquitous contaminants in silicon, is believed to participate in many complex defects (e.g. Cu-S. Cu-Se, Cu-Te, Cu-Zn, Cu-In, Cu-Cu pairs). However, many of the assignments in the literature have to be considered as doubtful and we expect to provide some definitive results regarding these identifications. With the isotopes ^{61}Cu (3.4 h) and ^{64}Cu(12.7 h) Cu related complexes can be formed and the decay time of the related lines will immediately give information on the number of Cu atoms involved.

The microscopic structure of a copper related luminescence center in silicon has recently been proposed to be a single Cu on a bond-centered position [23] contrary to former models of a Cu-Cu-pair defect [24]. Experimental evidence so far is more in favor of a Cu-Cu-pair. Here, the final proof could come from photoluminescence measurements on Si samples implanted with radioactive Cu where the decay of the luminescence, in case of a single Cu atom, should follow either the half-life of the isotope or vanish twice as fast in case of a pair-defect.

Transition metals are known to form deep levels in the band gap of Si, therefore complementary experiments with DLTS using the isotopes ^{61}Cu (3.37 h) and ^{64}Cu (12.7 h) will help to get a more complete picture of the electronic behavior of these elements. Using the long-lived isotope ^{56}Co (79 d) results have been obtained that are not yet fully understood [4]. We cannot exclude that the long-term stability of the samples may be a problem in this case. To overcome this problem, we intend to perform the experiments with ^{55}Co (18 h).

Identification of alkali and alkaline earth defects in silicon and germanium:

Alkali and alkaline earth metals are presumably major contaminants in Si and Ge. The concentrations, diffusion properties, and solid solubility of most of these elements have been derived from chemical analysis. Much less is known on their electrical and optical properties and their interaction with other defects. Deep levels are only reported for Li [25] but there exist hints for the complex formation with 5d elements and the electrical activation of isolated atoms by electron irradiation. The application of radioactive isotopes will allow to unambiguously determine the impurity concentration and to identify electrical and optical properties of these elements. Possible relevant isotopes are: ^{24}Ne; ^{28}Mg; ^{42}K; ^{43,44}Sc; ^{43}Ar; ^{47}Ca; ^{66}Ni; ^{66}Ge; ^{72}Zn; ^{73}Ga; ^{83}Sr; and ^{129}Ba. Useful half-lives mainly occur at the neutron rich side, i.e. one has to start with the corresponding noble gas element. In many cases (up to about mass 50) recoil energies during the decay can be rather large (several 100eV) which may

result in changes of the lattice site during the decay. We propose experiments involving alkali or alkaline elements themselves, the host element Si or Ge, and commonly used dopants. This would allow to identify alkali or alkaline earths correlated defects, to determine their electrical and optical behavior, and to study their complex formation with other defects.

3.2 III-V semiconductors:

Acceptors and donor states in GaN:

GaN and its alloys with AlN and InN are one of the most interesting classes of materials in actual semiconductor research due to their potential as optoelectronic devices like high efficiency LED and laser diodes in the blue and UV region [26]. Ion implantation into crystalline compound semiconductors is one of the main methods for selective introduction of dopant atoms. It offers control on concentration, depth and lateral distribution of the dopants. However, the use of ion implantation is always accompanied by undesirable structural damage to the crystal. The initial damage level depends on many factors such as substrate temperature, mass and energy of the implanted ion, dose and dose rate. Even under optimum implantation conditions thermal annealing is necessary to achieve electrical activation of the implanted dopant atoms. The understanding and control of the implantation induced defects and their interaction during the annealing process may become crucial for the realization of advanced devices based on GaN.

Especially p-type conductivity is difficult to obtain in GaN. Experiments reported in the literature on GaN doped with Mg during growth show that thermal annealing in hydrogen free atmosphere can electrically activate Mg acceptors [27]. These observations lead to the conclusion, that hydrogen is involved in the problems to achieve p-type GaN. It has been shown by PAC experiments that indeed hydrogen interacts with Cd acceptors in GaN [28]. However, the break up of the formed pairs occurs in a temperature range much lower than the temperatures needed to achieve electrical activation of dopants. Evidently there are additional defects in the material that interact with the dopants.

The electrical activation of implanted dopants in GaN is more complex compared to other III-V compounds since this material has both strong inter-atomic bonding demanding high annealing temperatures, and a constituent, N, having high vapor pressure. Techniques must be found which provide the high temperatures needed for crystal recovery while avoiding N loss from the GaN crystal which results in defects deteriorating the electrical properties of the material. It has been shown that annealing under a pressure of 10 kbar of N_2 after Zn implantation leads to an increase of a PL signal attributed to Zn [29]. However in materials that are so little characterized it cannot be excluded that the observed signal is related to other defects still present in the material.

An imperative prerequisite for these implantation studies is therefore the unambiguous identification of the PL and DLTS signals related to potential doping atoms in GaN. In this still highly defective material, however, this is difficult and for the case of PL mostly still based on the pioneering work of Pankove and Hutchby [30]. Using the isotope ^{111}Ag/^{111}Cd, we have demonstrated [31] the identification of the PL transitions related to Ag and Cd, and that about 30% of the Cd atoms act as acceptor.

For the case of the acceptor Zn, we propose PL and DLTS experiments using ^{67}Ga (78 h) decaying to ^{67}Zn to clearly identify the Zn related acceptor state. Here, the parent Ga will be

incorporated into a Ga lattice site forcing the decay product Zn onto the Ga lattice site. To determine the efficiency of the activation of implanted Zn ions, in a second experiment ^{69}Zn (13.8 h)will be implanted and the efficiency of the formation of the acceptor state and potential other Zn related states can be followed during its decay to ^{69}Ga. Here only the Zn atoms residing on unperturbed Ga lattice sites will give rise to the Zn acceptor levels identified above. All other Zn related states will give rise to new lines that also decay with the half-life of the implanted ^{69}Zn isotope.

An additional topic is the study of the states related to amphoteric Group IV dopants like Ge and Sn. They can either act as donor or acceptor depending if they occupy group III or group V lattice sites. A radioactive isotope for this study is ^{73}Ga which will occupy Ga sites after implantation. After its decay to ^{73}Ge, a donor state should be observable by PL and DLTS. Using ^{110}Sn decaying to the acceptor ^{110}Cd, PL and DLTS experiments should reveal if Sn forms an acceptor state in GaN.

Up to now, almost no reliable information is available on the electronic states of deep levels in GaN which can limit the carrier lifetime and their diffusion length. In a first experiment, we have shown that ^{191}Pt introduces a two new near-infrared transitions in GaN [32]. Using the radioactive isotopes ^{191}Hg, ^{192}Hg, and ^{193}Hg, the states of the decay products Au and Pt can be determined.

Since the residual shallow doping level in GaN is relatively high, the sensitivity of DLTS will be relatively low. Hence, implantation fluences in the order of 10^{12} cm^{-2} are expected to be necessary in contrast to typical fluences in the order of 10^{10} cm^{-2} used up to now for experiments in Si and SiC. These higher fluences will be available at ISOLDE only.

Antisite defects in GaAs and GaN:

An important class of intrinsic defects in a compound semiconductor of type AB like GaAs are antisites where an A atom is placed on a B site (A_B) or vice versa. Such defects might form during non-stochiometric crystal growth conditions or during particle irradiation and it is an important topic to know the energy levels of these defects. It is still an open question what the levels of the Ga_{As} antisite in GaAs are. From valence arguments Ga_{As} should act as a double acceptor. In agreement with this GaAs grown from a Ga rich melt is often p-type with an acceptor state at 78 meV and exhibits a PL emission at 1.441 eV [33]. Therefore this level and this emission is often assigned to the Ga_{As} antisite [34], however, there is also evidence that this level is due to boron on As sites [35]. A unique way to create Ga_{As} antisite defects in GaAs in a controlled way and to avoid the introduction of any other defect during the production process is the transmutation of radioactive ^{71}As (64 h) to stable ^{71}Ga.

In a first set of experiments [36,37] we have shown, that none of the PL transitions described in the literature is observed after the transmutation from $^{71}As_{As}$ to $^{71}Ga_{As}$. Therefore, we can exclude that any of these defect lines is caused by an isolated Ga_{As} antisite. Even more puzzling is the fact, that we do not observe any new PL line appearing between 0.9 eV and 1.515 eV during the population of the Ga_{As} sites (see Fig. 2). One possible explanation is that the emission related to Ga_{As} is very weak or very broad and therefore difficult to discriminate against the background due to contamination present in the sample, mainly C and Cu.

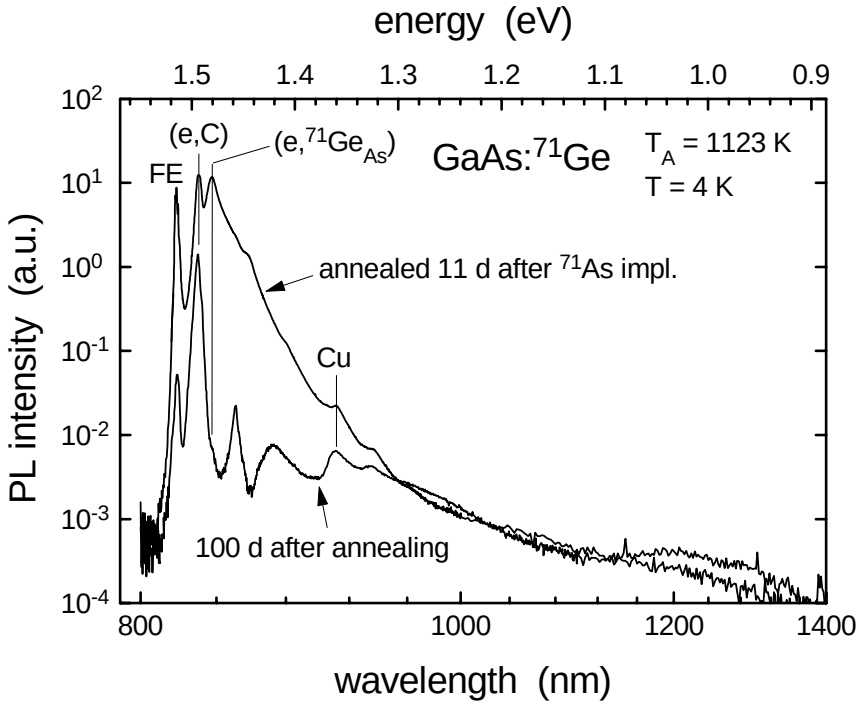

Fig. 2: *PL spectra measured at GaAs annealed at 1123 K 11 d after ^{71}As implantation and 100 d after the annealing treatment.*

Therefore we intend to repeat this experiment by using MBE grown GaAs layers with a background level of contamination much lower as in the previous experiments. If on the other hand Ga_{As} antisites indeed do not produce any shallow levels visible by PL it might form deep levels in the band gap. Since, in contrast to PL, DLTS is expected to detect any deep level, this experiment should also be done with radiotracer-DLTS to identify the electrical properties of the Ga antisite defect.

Almost nothing is known about the Ga_N antisite in GaN. Theoretical investigations of Jenkins *et al.* calculate the Ga_N antisite level at 0.7 eV above the valence band [38], Boguslawski *et al.* [39] calculate the Ga_N antisite level at around E_V +1.4 eV, and Neugebauer *et al.* state that antisites in Ga_N are energetically less favorable [40] than in other III-V semiconductors. In a first experiment using ^{71}As [41], we found no PL transitions related to the Ga_N antisite, but this experiment must be repeated and completed by DLTS and Hall effect measurements. A non-existent PL transition does not exclude the existence of the antisite, because the related transitions could be non-radiating.

3.3 II-VI semiconductors

p-type doping of ZnSe:

With exception of CdTe, most II-VI semiconductors can be doped only either p-type (ZnTe) or n-type (e.g. ZnSe, ZnO) with high efficiency. In particular, the problem of achieving p-type conductivity in ZnSe has been a subject of intense research aiming for the fabrication of blue laser-diodes. However, the identification of acceptor levels in ZnSe is often speculative and sometimes contradictory [42,43].

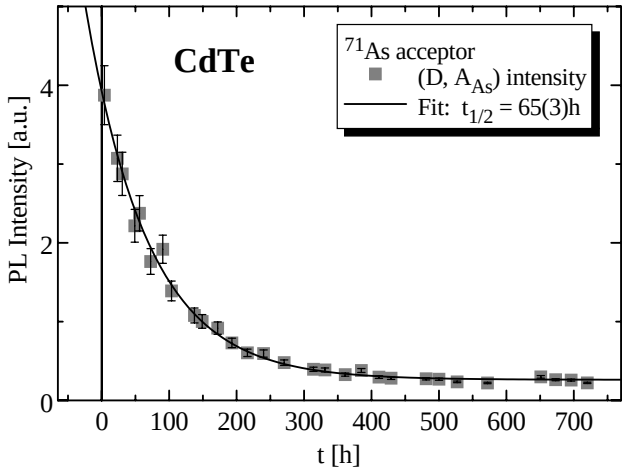

Fig. 3: *Intensity of the (D, AAs) transition, caused by radioactive ^{71}As isotopes in CdTe, as a function of time.*

While the determination of the Ag acceptor-levels in CdTe and ZnTe was successful by using the radioactive ^{111}Ag isotope ($t_{1/2}$ = 7.5d) for PL spectroscopy [44], no PL signal related to an Ag-acceptor could be observed after implantation of ^{111}Ag into ZnSe. This observation indicates that during annealing, which is necessary for the incorporation of the implanted ions and for the removal of radiation damage, the group I element Ag either is not incorporated within the Zn-sublattice or it forms a an electrically inactive complex. In order to force the correct incorporation of Ag, ^{107}Cd isotopes ($t_{1/2}$ = 6.5 h) can be used which are isoelectronic to the Zn host and, therefore, are expected to occupy undisturbed Zn-sites after annealing. After the decay of ^{107}Cd to ^{107}Ag, the detection of Ag_{Zn} acceptors should be possible. The need to perform PL measurements on-site at ISOLDE is given by both the short half-life of ^{107}Cd and the observation of an increasing PL-signal. A similar behavior as for the group I acceptor Ag is observed for the group V acceptor As, which resides in the chalcogenide sublattice: By means of ion implanted ^{71}As isotopes ($t_{1/2}$ = 65h), the unambiguous identification of the As-acceptor in CdTe was achieved (see Fig. 3). In contrast, a ^{71}As related PL-signal was not observable in ZnSe [45]. Here, the implantation of short-lived ^{73}Se isotopes ($t_{1/2}$ = 7.1h) can constitute a possibility to produce As_{Se} acceptors by radioactive transmutation.

The detailed character of the temporal development of the PL signals depends on the annealing behavior of the parent and the daughter isotope. On the one hand, this fact can be used to extract information on the annealing behavior of the involved elements [45]. On the other hand, the necessity to start the PL measurements immediately after implantation can be given even for rather long-lived isotopes. For example, the identification of Au-related PL signals in CdTe was successful after implantation of ^{197}Hg ($t_{1/2}$ = 64 h, daughter isotope ^{197}Au) [46], because the annealing time turned out to be the time-zero for the intensity development of the Au-related PL. After implantation of ^{197}Hg into ZnTe, however, the time constant describing the increase of PL intensity that is caused by Au-acceptors could not be determined with sufficient accuracy, because the Au atoms already present at annealing time did contribute to the Au-related PL, so that only a relatively small change on the top of an offset was observed during the sequence of PL measurements. In this case, the time-zero for the development of the PL signals of interest is the implantation time, hence the possibility of on-

site measurements minimizing the delay between implantation and measurements is highly beneficial.

p-type doping of ZnO:

Although GaN has been the material of choice for short wavelength optoelectronic devices (LED, laser diodes) in recent years, the report this year of laser action in ZnO has opened up a major new technology field [47]. ZnO has a direct band gap of 3.5 eV similar to GaN. In contrast to GaN which can only be produced by epitaxial procedures, bulk ZnO crystals of good quality can now be produced, but ZnO is still one of the most poorly understood materials with respect to impurities, defects, and associated energy levels. For example, ZnO is typically n-type, and general agreement as to the chemical identity of the dominant donor impurity (interstitial Zn, oxygen vacancies, or hydrogen) is not yet established [48]. It is still difficult to produce p-type material in a reproducible fashion, and this remains an important problem regarding further technological development, i.e. building diodes. As in the case of donors, the identity of the acceptors found in as-produced ZnO is not yet proven. A review of the main issues regarding ZnO has been published by D.C. Look [49].

The experiments we propose will deploy the proven power of PL and DLTS with radioactive isotopes for the study of selected impurity and defect systems in ZnO. PL is very well developed for the study of ZnO [50] and, recently, Schottky contacts (necessary for the performance of DLTS measurements) have been produced, so the necessary basic requirements for successful experiments are in place. In the following, reference is made principally to PL, as there are as yet very few reports of capacitance transients or DLTS measurements on ZnO in the literature.

The focus of the first cycle of experiments will be on the properties of impurities similar to the group II host atom Zn. The objectives are (i) the identification of the PL and DLTS signatures of selected donor, acceptor and neutral impurity centers, and (ii) the study of the interactions of implanted atoms with the host crystal. We have chosen the following set of isotopes in order to provide a balanced and systematic analysis of the impurities involved.

$^{62}Zn \rightarrow Cu \rightarrow Ni$

Implanted ^{62}Zn impurity atoms are expected to preferentially occupy Zn sites in the ZnO crystal, and the ^{62}Zn decay sequence will produce Cu_{Zn} and Ni_{Zn} acceptor centers. Our experiments will be used to verify the identification of luminescence and DLTS bands related to Cu_{Zn} and Ni_{Zn}.

$^{72}Zn \rightarrow Ga \rightarrow Ge$

In this case, the implanted ^{72}Zn decay sequence will produce the Ga_{Zn} and Ge_{Zn} donor centers. As for case (i) above, the luminescence bands and DLTS levels will be studied for the two donor species. The choice of ^{62}Zn and ^{72}Zn provides a perfect means of establishing the key data for both donor and acceptor centers associated with the Zn sites in the ZnO crystal.

$^{67}Cu \rightarrow Zn$

The ^{67}Cu isotope is selected because it provides the reverse transformation of Cu to Zn compared to the first case listed above. In particular, any Cu that occupies interstitial sites will result in the formation of interstitial Zn (Zn_i) and opens the possibility to identify Zn_I which is discussed as one of the dominant donors in as-grown ZnO.

$^{66}Ga \rightarrow Zn$

In this case, the parent isotope ^{66}Ga decays to the host element Zn, enabling the properties of implanted Ga centers to be studied and compared to the Ga_{Zn} centers produced from the decay of ^{72}Zn described in the second case above.

$^{104}Cd \rightarrow Ag \rightarrow Pd$

The ^{104}Cd atoms are expected to occupy Zn sites producing neutral (isoelectronic) Cd_{Zn} centers. The decay to Ag and then to Pd should produce the Ag_{Zn} and Pd_{Zn} acceptors which will then be studied in detail and their properties compared to those of Cu_{Zn} and Ni_{Zn}.

$^{192}Hg \rightarrow Au \rightarrow Pt$

In this case, the implanted ^{192}Hg is again expected to occupy Zn sites and produce an isoelectronic centre which decays to the acceptors Au and Pt. Here we will have a tool of studying the potential acceptor species ZnO:Au (for comparison with ZnO:Cu and ZnO:Ag) and ZnO:Pt (for comparison with ZnO:Pd and ZnO:Ni).

In summary, the experiments we propose will provide a broad set of examples of isoelectronic, donor and acceptor impurity centers in ZnO. Variations in the interactions of the different implanted elements with the host crystal will also be determined. The results will provide clear identifications of luminescence and DLTS levels and will thereby produce much needed unambiguous experimental data for developing the study and understanding of impurities and defects in ZnO which are prerequisites for the development of new devices based on this material.

3.4 Deep states in SiC

Silicon carbide is one of the wide-gap semiconductors that attract great interest in current semiconductor research [51]. SiC is expected to replace traditional semiconductors like Si or GaAs in some specialized areas like high-power or high-frequency devices [52]. Because of its extreme chemical and thermal stability, it is expected to open up new semiconductor applications in rough environments and high-temperature electronics.

The proposed experiments on group III elements in SiC were motivated by reports in the literature, predicting a Ga-related deep center in the band gap of silicon carbide. A shallow acceptor state produced by Ga residing on Si lattice sites [53] was detected by Hall effect experiments and admittance-spectroscopy at 0.30 eV above the valence band edge in 6H-SiC [54]. In the case of B, the existence of both a deep and a shallow state is well established and explained by two different structural configurations [55]. There is a clear conclusion from magnetic resonance experiments [56] that the group III elements B, Al, and Ga tend to form - in addition to the expected shallow state - a complex involving a vacancy which acts as a deep center. Such a deep level, however, has not been detected except for boron. First experiments with the transmutation of ^{67}Ga $\rightarrow$ Zn [57] did not reveal any evidence for a deep level of Ga in the accessible energy range.

A repetition of this experiment at ISOLDE with a higher fluence of ^{67}Ga than in the previous recoil implantation experiments will help to reduce the lower limit for the fraction of Ga involved in such a state; at present, this limit is not satisfactory.

4. Experimental requirements

Depending on the experiment the collections will either be performed in the solid state physics implantation chamber connected to the GLM or GHM beam line, or if higher energies are needed at the high voltage platform with an implantation energy of up to 260 keV. The GLM and GHM beam lines are both equipped with a beam sweep system necessary to achieve a homogeneous doping of the samples. The solid state physics chamber allows the implantation of up to 10 samples at variable temperatures without breaking the vacuum.

The sample preparation before (cutting, cleaning, etching etc.) and after the implantation (thermal annealing, electrical contacts etc.) will be done in the solid state physics laboratory in building 115 at ISOLDE.

To set up photoluminescence experiments at CERN a number of requirements have to be met to be able to comply with CERN safety standard. This requires a laser laboratory in which we can work with lasers of class 3b, radioactivity and liquid helium. The solid state physics groups funded by the German Minister for Research (BMBF) have raised the money (465 000 DM) for a state-of-the-art photoluminescence spectrometer to be installed at CERN. This spectrometer is already delivered to CERN and can be installed immediately when the necessary laboratory is provided.

The DLTS and Hall effect measurements consist of less delicate equipment and can be brought to CERN for the experiments. Nevertheless, they need space to be temporarily installed, preferably in the same laboratory where the PL spectrometer is installed.

The laboratory should satisfy the following basic requirements:

- closed laboratory of at least 40 m^2 surface for PL, DLTS etc. or 2 labs of at least 20 m^2 each, which should have no windows or light-tide Venetian blinds. The labs should have an area with sufficient height (about 4 m) to allow the use of standard liquid helium dewars and transfer lines;
- stable non vibrating floor for optical installations;
- water cooling for the lasers;
- sufficient electrical power outlets (3 phases, at least 24 A per phase) to power the lasers;
- laser safety installation such as curtains , restricted access, warning lamps;
- fulfill the requirements to use open radioactive sources;
- helium gas recovery line;
- exhaust line for vacuum pumps;
- air conditioning;
- ethernet connection to the CERN network.

For the off-line Mößbauer experiments the same laboratory can be used. Here the stable floor without sources of vibrations is as necessary as for the optical setup. In view of the ISOLDE consolidation project and the almost non-existing space resources in the ISOLDE hall, a laboratory able to hold all solid-state physics and life sciences experiments permanently

or temporarily installed at CERN (perturbed angular correlation, emission channeling etc.) would be highly desirable.

To optimize the transmission into GHM or GLM about 2 hours of stable beam is required before the experiment, for experiments using the high voltage platform about 1 shift of stable beam is required.

5. Beam time request

Depending on the system under study and the used spectroscopic technique, the total required number of implanted ions ranges from 10^{10} up to 10^{12} ions per sample. Taking into account the yields for the required isotopes near to stability, implantation times between minutes and a few hours per sample are required. Most of the beam-times for a certain target-ion-source combination can and should be shared with other experiments.

Since the preparation of the experimental setup takes about one day each beam time should run at least 3 days to be able to perform several different measurement series during one beam time. Depending on the number of users with whom we can share the beam this means between 4 and 9 shifts per target. For a reasonable working progress each target should be available at least once per year.

We request a total of 40 shifts for the years 2002 and 2003 for the following isotopes:

Isotope	**half-live**	**Target**	**Ion source**	**Energy**
^{24}Ne	3.4 min	SiC,MgO,UC_2	Plasma	260 keV
^{28}Mg	21.0 h	UC_2, ThC_2	hot plasma	260 keV
^{42}K	12.4 h	Ti, Ta, UC_2	W surface	260 keV
^{43}Ar	5.4 min	ThC_2, UC_2, TiO	plasma	260 keV
^{43}Sc	3.9 h	Ti	W surface	260 keV
^{44}Sc	3.9 h, 58.6 h	Ti	W surface	260 keV
^{47}Ca	4.5 d	UC_2, ThC_2	W surface	260 keV
^{55}Co	17.5 h	Nb or Ta *	hot plasma	260 keV
^{61}Cu	3.4 h	ZrO_2, UC_2	Laser	60 keV, 260 keV
^{64}Cu	12.7 h	UC_2	Laser	60 keV, 260 keV
^{66}Ni	55 h	UC_2	Laser	260 keV
^{66}Ge	2.3 h	ZrO	hot plasma	260 keV

^{66}Ga	9.4 h	Nb or ZrO_2	hot plasma	60 keV
^{67}Ga	78.3 h	Nb or ZrO_2	hot plasma	60 keV, 260 keV
^{73}Ga	4.9 h	UC_2, ThC_2	W surface, Laser	60 keV
^{62}Zn	9.2 h	Nb	hot plasma	60 keV
^{69}Zn	13.8 h	UC_2, ThC_2	Laser	60 keV, 260 keV
^{72}Zn	46.5 h	UC_2, ThC_2	hot plasma, Laser	60 keV
^{71}As	64 h	Nb	hot plasma	260 keV
^{73}Se	7.1 h	ZrO_2	hot plasma	60 keV, 260 keV
^{83}Sr	32.4 h	Nb	W surface	260 keV
^{104}Cd	58 min	Sn molten	plasma heated line	60 keV
^{107}Cd	6.5 h	Sn molten	plasma heated line	60 keV, 260 keV
^{110}Sn	4 h	LaC_x	Laser	60 keV, 260 keV
^{129}Ba	2.2 h	Ta, molten La	W surface	260 keV
^{189}Hg	11 h (Pt)	molten Pb	hot plasma	60 keV, 260 keV
^{191}Hg	3.2 h (Au)	molten Pb	hot plasma	60 keV, 260 keV
^{192}Hg	5 h (Au)	molten Pb	hot plasma	60 keV, 260 keV
^{193}Hg	3.5 h (Pt)	molten Pb	hot plasma	60 keV, 260 keV

(For several isotopes, the laser ion source could replace the hot plasma ion source)

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